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Role of Charge Localization in the Basic High Temperature Superconductivity Mechanism

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Abstract

The elucidation of the mechanism of superconductivity in the cuprates is one of the most important problems in condensed matter physics. We have developed an array of experimental and theoretical tools for studying and understanding the "mesoscale" structure (structure on length scales intermediate between the crystalline lattice constant and the macroscopic length scales characteristic of "averaging" probes such as diffraction, transport, and thermodynamic experiments). Advances in understanding these problems hold the promise of making a broad range of materials with novel and technologically important properties avail-able for exploitation. We describe experimental tools we developed to measure the properties of materials in this mesoscale regime; analytic techniques we developed to quantify and model mesoscale structure and complexity, and to determine how they control material functionality; and important advances in experimental understanding of detailed relationships between charge ordering and functional properties in several members of a very important class of electronic materials.

Background and Research Objectives

The majority of early studies of the cuprates were guided by our traditional understanding of solids based on the picture electrons interacting with periodic arrays of atoms in lattice positions given by the crystal structure. However, several different kinds of measurements have recently shown that this picture is often not correct in systems where the electrons are strongly correlated, most particularly in the cuprates. Atoms are often displaced from their ideal positions, and these displacements can have local correlations of several to hundreds of lattice constants. This structure may persist for times long compared to a phonon vibration time, and the local excitations may be highly nonlinear. These effects are present even in a material that is nominally free of the usual defects, such as vacancies, interstitials, and substitutional impurities. A further very important phenomenon, almost certainly related to these local structure effects, is the tendency of added charged carriers to distribute inhomogeneously. Recent studies, including our own, in various lanthanum cuprates demonstrate that many of the doped charge carriers are localized in the CuO_2 planes. This suggests strong charge-lattice interactions, and this can be expected to have important ramifications for the behavior of the remaining itinerant charge carriers. The observation of static stripe ordering in Sr doped lanthanum nickelate (isostructural with lanthanum cuprate) motivated the search for similar structures in the cuprates [1-5]. This led to the discovery of static charge stripes in rare earth co-doped lanthanum cuprate through neutron diffraction measurements [6]. This leads one to con-

sider that charge-stripe formation may be a rather general phenomenon in cuprates. Tranquada *et al.,* argued [7] that the stripes are only observed as static in the case some feature of the lattice pins the stripes, otherwise they are dynamic. Charge ordering will fundamentally affect materials properties and may help explain the novel normal state properties of the HTC cuprates, and very importantly, localized charge and ordered arrays of such states could provide the basis for the superconducting pairing mechanism.

There is a substantial literature demonstrating that locally, crystal structure deviates from the global crystallographic structure observed by diffraction probes. Using NMR and NQR studies we demonstrated the existence of local inhomogeneities in crystal structure that are not due to distortions arising from proximity to dopants but are the intrinsic response of the CuO_2 planes to the presence of doped holes. The local structure studies provide a time snapshot of structure whether periodic or not. It is well suited to studying the response of the lattice to charge ordering that is dynamic and not periodically ordered. It now appears likely that many of the local structural effects that have been observed reflect the response of the lattice to dynamic charge ordering.

Our goals, then, were to develop experimental and theoretical techniques that are effective for understanding of the complex spatially inhomogeneous and temporally dynamic states found in the high temperature superconductors, and to apply these techniques to prototypical superconducting cuprates and to related materials. From these studies we sought to gain improved understanding of the character of and the mechanisms responsible for the formation of these complex electronic states, and to illuminate their role in high temperature superconductivity.

Importance to LANL's Science and Technology Base and National R&D Needs

Los Alamos must not only retain but stretch its world-class competency to systematically control tunable classes of correlated electron materials for targeted technology issues. With dwindling resources this is clearly becoming the approach to R&D on many-body electronic materials nationally. By history, reputation and infrastructure, Los Alamos is the ideal environment to lead this national agenda, involving industry, universities and national laboratories. Of the major national laboratories, Los Alamos in unique in both is breadth and depth in correlated electron materials, including high temperature superconductors. No other institution has the combined abilities to develop new materials, characterize them and carry out theoretical modeling at all levels. Our leadership role in the nation and the world is even more crucial as the major industrial laboratories dismantle their basic and applied scientific research efforts. (Corporations reason that although this research is vital to the nation's economy, it does not benefit primarily their own com-

pany's bottom line.) Correlated electron physics is a scientifically vital area of research that continues to excite the imagination and challenge our physical intuition, and at the same time opens new avenues of electronics applications for industrial and defense technologies where the anticipated U.S. annual product is in the multi-billion dollar range. This thrust has helped to maintain and stretch Los Alamos' competency in advanced and nuclear materials, in particular, those in which electronic correlations play a dominant role, and provide the scientific basis for ideas that we and the nation will "mine" for technologies of the next century. We have laid the path for working synergistically with industry, agencies, universities and other national labs to develop a focus for (a) identifying key classes of novel electronic materials characteristics defined by industrial and weapons requirements and (b) controlling the fundamental science issues for optimization of the identified characteristics. We have shown the value of developing new theoretical and modeling tools and strategies, and working closely with synthesis and characterization programs to "close the loop," which is key to predictive control for synthesis-microstructure-property relations. This program has contributed to a fundamental understanding of high temperature superconductivity, enhancing efforts to develop high current superconductors at high temperatures and magnetic fields. This program utilized and provided scientific leadership and support for unique Laboratory facilities (NHMFL, MLNSC, ACL) and leveraged existing DOE/OBES, LDRD, UC, etc. programs. In addition we have collaborated with programs in CMS, STC, CNLS and with UC to provide a national focus through collaborations, visitors, etc., involving industry, universities and other national laboratories.

Scientific Approach and Accomplishments

The core of our scientific approach involves the combined application of experimental and theoretical studies to the key issues identified above. We have experimentally studied the response of cuprates to the addition of in-plane impurities, carefully explored the experimental consequences of charge stripe order, developed techniques for the study of local structure and applied these to cuprates to detect the structural signatures of charge stripes.

An effective strategy for studying the behavior of both superconducting state and normal state charged carriers in the cuprates is to study their response to the addition of impurities and defects that perturb the carriers. To understand the normal state magnetism of the cuprates we have examined the effect of adding defects in the electronically active CuO_2 planes, in comparison to more weakly interacting out-of-plane impurities. These impurity studies were applied to both the normal and superconducting states. An-

other thrust of our experimental work has been to perform structural studies which avoid any assumption of lattice periodicity. This enables determination of local structures that deviate from the average underlying crystal structure. In this work we have obtained a detailed understanding of the local structure of the prototypical lanthanum cuprate high T_c superconductor; this foundation has enabled a successful search for the structural signatures of dynamic charge-stripe order in this very important cuprate superconductor. It is now widely recognized that charge-stripe order is a common response of two-dimensional transition metal oxides to the addition of low densities of charged carriers. The successful observation of static charge stripe order in certain lanthanum cuprate compounds indicates that charge stripes may play an important role in determining the normal state properties of the cuprates as well as the mechanism of superconductivity. In addition to the structural work just mentioned, we have studied the magnetic properties of a lanthanum nickelate, a transition metal oxide isostructural to lanthanum cuprate that exhibits robust charge-stripe order. Recently we have begun studies of the more complex static stripe-ordered lanthanum cuprate compound.

One thrust of this effort has been the study of Li-doped La_2CuO_4. La_2CuO_4, the prototypic single-layer cuprate, can be chemically hole-doped in two ways: out-of-plane Sr doping and in-plane Li-doping. In both cases one hole is added to the CuO_2-planes per dopant atom. In the Li case, the doped material never becomes metallic or superconducting. It is therefore remarkable that Li-doping in other respects is identical to Sr-doping. Namely, the tetragonal-orthorhombic transition temperature and the copper-oxygen plaquette size as a function of doping concentration are independent of which dopant is used (see Pub. 9). ^{139}La nuclear quadrupole resonance measurements in lightly doped $La_2Cu_{1-x}Li_xO_4$ showed that both the magnetic ordering temperature and the details of the evolution of the ordered magnetic moment with temperature is remarkably insensitive to placing the impurities into the CuO_2-planes (see Pub. 16). We also found that two unusual magnetic features characteristic of antiferromagnetically ordered lanthanum cuprate behave very similarly in the two systems (see Figure 1). These are the freezing of spin-degrees of freedom, accompanied at slightly higher temperatures by the abrupt recovery of the sublattice magnetization almost to values found in the undoped antiferromagnet. These results indicate that even in the low doping regime, the added holes form collective structures whose behavior determines the magnetic properties (see Pubs. 16-18).

We have studied the magnetism of charge-stripe ordered two dimensional transition metal oxide $La_{5/3}Sr_{1/3}NiO_4$ (isostructural to La_2CuO_4; see Pub. 20). In our single crystal ^{139}La NMR study we were able to observe the two magnetically distinct sites resulting

from charge-ordering; the first located in the charged stripe, and the second in the intervening hole-free domains. We observed a marked sensitivity of the magnetic ordering temperature to the measurement time scale (see Figure 2). This sensitivity is a hallmark of glassy systems, and provides clear evidence that the charge-stripes form into a glass-like solid. We also discovered a pronounced and unusual spin disorder arising from stripe defects. It is quite interesting that this disorder persists to low temperature where charge order becomes very good.

Pair distribution function (PDF) analysis of neutron diffraction data provides structural information without any assumption of lattice periodicity. Thus it is able to determine local structure which deviates from the average (periodic) structure. These techniques provided some of the earliest evidence for inhomogeneities in the cuprates, and it now appears likely that these observations foreshadowed the observation charge-stripe order. Because the PDF technique is sensitive to dynamic fluctuating structures, it will be sensitive to dynamic stripes. We have written an extensive review (Pub. 34) of the experimental situation regarding the observation of lattice effects and local structure in high-temperature superconductors as of 1996 describing the state of the art before they were recognized to be arising from stripes. Publication 29 gives a detailed description of how the PDF analysis of neutron scattering data is performed and local structural information is obtained. One of the successes of the work reported here has been the development of the sophisticated data collection, analysis and modeling techniques which has made possible the observation of dynamic stripes in the cuprates as well as important advances in studies of other transition metal oxides such as the manganites. This paper gives a detailed account of technical aspects of how these measurements are performed.

We have performed an in-depth study of the variation in the local structure with doping (Pub. 26) to observe dynamic stripes in superconducting compositions of high-temperature superconductors. The charge stripes are coupled to the octahedral tilts in these materials which we can measure locally. If small domains of stripe order exist and are fluctuating we should see tilt disorder at intermediate doping levels. We show evidence that this is indeed the case; there appear to be a distribution of tilt amplitudes, and possibly directions, at intermediate doping levels in $La_{2-x}Sr_xCuO_4$ which supports the view that there is an inhomogeneous distribution of charge in the CuO_2-planes for compositions that are superconducting.

We have made a thorough study of the effect of in-plane impurities on the superconducting state of the high T_c cuprates (see Pubs 22-23). We have discovered that when Ni impurities are doped in high temperature superconductor Bi2212, thermal conductivity displays a sharp drop at a temperature of $T^* = 200$ mK (see Figure 3). The sample just

above T* is already in a superconducting state, having undergone the high temperature superconducting transition around T_c = 80 K. Superconducting electrons do not contribute to thermal transport; only normal electrons remaining in the sample below T_c can carry heat. We propose that additional reduction of thermal conductivity at T* reflects the sharp reduction of the number of normal electrons in the sample due to a change in the superconducting order parameter.

In collaboration with M. Salkola (STI), J. R. Schrieffer (NHMFL), and D. Scalapino (UCSB), we theoretically investigated these impurity bound states in d-wave superconductors (Pubs. 42 and 43). Impurity scattering in unconventional superconductors suppresses the critical temperature. The physics of the impurities beyond simple lifetime considerations turns out be very interesting. The real space shape of the impurity states is highly anisotropic, and exhibits a four-fold symmetry coming from the anisotropic energy gap of superconducting electrons. We calculated both the energy and the real space shape of impurity bound states.

We introduced the notion of marginal stability of a d-wave superconductor in the presence of external perturbations, such as magnetic impurities or external magnetic fields. We argued that quasiparticles at the nodes of the initial d-wave state are susceptible to secondary pairing in the id_{xy} channel to maximize the condensate energy.

We found that impurity scattering not only generates a scattering lifetime but also can produce a secondary component of the order parameter in d-wave superconductors. Specifically, spin orbit scattering can generate a complex id_{xy} component of the gap near the impurity with d_{xy} symmetry and a phase that is shifted by 90° with respect to the original d_{x2-y2} component. This secondary component forms around the magnetic impurity as a patch the size of the coherence length. When the impurity concentration on the order of few percent, these patches begin to overlap, possibly leading to the formation of a coherent $d_{x2-y2} + id_{xy}$ state in the sample (see Figure 4). Together with the initial d-wave gap this effect will lead to a locally "quasi gapped" spectrum near impurity sites (see Figure). At low temperatures these $d_{x2-y2} + id_{xy}$ patches can order to form a global $d_{x2-y2} + id_{xy}$ superconducting state. This state violates the parity P and time reversal T symmetries and is similar to the secondary superconducting transition observed in some heavy fermion materials. The experimentally observed second superconducting transition at 0.2 K in Ni-doped Bi2212 (discussed above) is the realization of this phase ordering transition.

We measured the thermal conductivity of insulating Bi2212 (Yttrium-doped Bi2212), to directly measure the lattice (phonon) contribution to thermal conductivity. The results indicate that most of the heat current in superconducting Bi2212 is carried by the electrons. Our preliminary measurements of the specific heat of Ni-doped Bi2212 in zero and

applied magnetic field (up to 9 T) between 50 mK and 2 K indicate possible spin-glass ordering of Ni impurity spins. This finding is in accord with the theory of this phenomenon proposed by A. Balatsky (Pub. 37).

As a cuprate dopant, Li is unique in that it is sufficiently soluble in La_2CuO_4 that one can form $La_2Cu_{0.5}Li_{0.5}O_4$ in which there is one hole per Cu, in this case the Li and Cu ions order periodically to form a superlattice (every other Cu site is periodically occupied by Cu then Li). Hence, this material is unique amongst cuprates in that one is able to study the magnetic behavior of the doped hole confined by the four Li neighbors and in the absence of the confounding contributions of the antiferromagnetically correlated copper moments. It turns out that this arrangement provides a prototypical realization of a regular lattice of so-called "Zhang-Rice singlets," the theoretical description of the charge carrier in the cuprates that is at the core of many models of high-T_c superconductivity. This enabled the first experimental examination of these theoretical ideas. Our experimental nuclear quadrupole resonance results revealed that the actual situation is much more complicated than the theorists had imagined (see Pub. 10). In particular we found that the energy of the excited state relative to the ground state is smaller than predicted by Zhang and Rice, and strikingly similar to the value of the effective exchange interaction J in the lightly doped cuprates. This suggests the involvement of some other physics which introduces low-lying states into the excitation spectrum of the doped hole state.

High magnetic field NMR studies were performed at the National High Magnetic Field Laboratory (NHMFL) to study the response of the spin pseudogap to high fields (Pub. 19). This study showed that the spin pseudogap is independent of field and demonstrates that the opening of the spin pseudogap and the onset of superconductivity are distinct events.

There is growing experimental evidence in cuprate and related transition metal oxides (nickelates, bismuthates, manganites, ...) from neutron and x-ray scattering, direct STM imaging, etc., for charge localization and mesoscopic charge ordering (with attendant glassy dynamics and inhomogeneous, filamentary transport). This intrinsically multiscale situation of nanoscale phase separation of charge and spin, leading to coexisting inhomogeneous metallic and magnetic patterns, is difficult to approach theoretically from a microscopic model. We have therefore (much as in traditional solids) approached the problem in two steps: (a) what forces can control the charge localization and mesoscopic ordering; and (b) what are the excitations (spin, charge and lattice fluctuations) associated with given mesoscopic patterns. This is important for their identification and functionality.

Concerning issue (a), we have shown that competing short- and long-range forces lead to rich varieties of charge ordering in antiferromagnetic spin backgrounds appropriate to layered cuprates. The short-range forces arise from disrupting the local spin order, from impurity pinning, and from polaronic-type coupling to local lattice distortions. (There are equivalent short-range forces upon doping in uniform charge-density-wave or Jahn-Teller broken-symmetry ground states.) The long-range forces arise from Coulomb interactions and from elastic interactions, which are especially strong in transition metal compounds. We have studied these various contributions with modeling techniques including semiclassical treatments of holes with long-range interactions, nonlinear shell models appropriate to polarizable perovskites, and nonlinear elasticity theory which self-consistently generates anisotropic long-range interactions. We have considered charged and neutral impurities, both in-plane and out-of-plane, relevant to experimental studies with Zn, Sr, Ni, O, Li, etc. The most important conclusion is the robustness of "stripe segments" of ordered holes, which are entirely consistent with available experiments but point to the need for far better "mesoscale" probes for further discrimination. The dynamics and melting of these patterns (both quantum and thermal) is under study. Preliminary results support glassiness and spatio-temporal intermittency, suggesting new dynamic experimental probes such as nonlinear transport and susceptibility, and dynamic inelastic neutron scattering, in addition to existing evidence for dynamics from differences between NMR, neutron, x-ray, and other data (see Pubs. 70,71, 73,74).

Concerning the excitations accompanying the nanoscale patterns, we have focused on two issues. First, we have used a combined real-space Hartree-Fock plus Random Phase approach in a multiband Peierls-Hubbard model to determine linear spin, charge and lattice vibrations. These will be well-defined excitations if their frequencies are fast compared to any slow evolution of the global charge-ordered pattern. We find very distinctive low-lying new "phonon" and "spin-wave" modes associated with the charged stripes and their boundaries with the magnetic background (i.e. modes strongly localized around the filamentary charge skeleton). Most interestingly, these novel lattice and spin excitations have similar energies, suggesting strongly resonant "magneto-elastic" coupling but acting very inhomogeneously, whose consequences for superconductivity are under study. Second, we have considered the coupling of in-plane magnetism with out-of-plane (c-axis) polarizability, arising from the sensitivity of apical oxygen ions to their local charge environment. This polarizability takes the form of nonadiabatic polaron tunneling with distinct signatures both structurally and optically, which is consistent with much experimental data, including oxygen isotope effects. Intriguingly, allowing for local buckling of in-plane oxygens (probably inhomogeneously from charge-ordering and impurities, as

discussed above) leads to strong coupling of in-plane and c-axis lattice vibrations. This produces a coupling of the c-axis polarizability to the in-plane magnetism, inhibiting permanent ferroelectric dipoles but using the slow quantum paraelectric fluctuations to enhance superconducting pairing in the plane. This resonant enhancement scenario for superconductivity is an appealing HTC mechanism, and we are continuing to study its ramifications (Pubs. 63, 65).

We have also used the above techniques to study charge localization, ordering and fluctuations in other transition metal oxides viewed as complementary to the cuprates - manganites, nickelates, and $La_2Cu_{0.5}Li_{0.5}O_4$. Comparisons with recent experimental data at Los Alamos, especially NMR (Hammel et al., discussed above) and inelastic phonon neutron scattering (McQueeney et al.) are very encouraging and give reliability checks for the more subtle case of the HTC cuprates (Pub. 69).

Cluster approximations (Pub. 44) to the electronic structure of La_2CuO_4 were used to determine the Cu nuclear quadrupole resonance (NQR) spectrum (Pub. 45) for comparison with the experimental efforts (Pub. 2) in MST-10. It was shown that the experimental spectrum is consistent with holes localized in either the CuO_2 plane or on oxygens in the interstices of the lattice. The electronic correlations which drive the localization in the plane were identified (Pub. 46) and a simple model presented which reproduces the first principles calculations. At minimum, a three-band model with an on-site repulsion on the Cu is necessary to describe this behavior, and it arises from a Kondo-like interaction between holes in the Cu3d and the O2p levels. It was furthermore shown that this interaction will bind a pair of holes in larger cluster models of the cuprates (Pub. 47).

A problem occurring in many materials near the metal-insulator transition is that the workhorse of first-principles band-structure theory, the local density approximation (LDA) of density functional theory (DFT), predicts metallic (delocalized) behavior when insulating (localized) behavior is actually observed. For example, the LDA predicts undoped La_2CuO_4 to be a metal, when in fact it is an antiferromagnetic insulator with a gap of some 2eV. Other examples of qualitatively incorrect predictions by the LDA include the heavy-fermion materials, other transition-metal oxides, and some actinide materials.

We found that cluster models of La_2CuO_4 studied with the LDA also overemphasize delocalization. The generalized-gradient-approximations (GGA) of DFT did not qualitatively improve the description. A hybrid theory (Pub. 48), which combines the full nonlocal "exact" exchange interaction with the LDA was shown to lead to marked improvement in the description of antiferromagnetically coupled sites in clusters. Semiquantitative agreement with experiment was found for the magnitude of the coupling constant in La_2CuO_4, $KNiF_3$, and K_2NiF_4. The magnitude of the unpaired spin population on

the metal site was in excellent agreement with experiment for La_2CuO_4. The theory (Pub. 49) was also used to study the antiferromagnetic interaction in the ladder material $SrCu_2O_3$ (Pub. 50).

Most theories of superconductivity are based on electrons interacting with a boson, such as a phonon or spin fluctuation. The interaction modifies the properties of the electron, forming a quasiparticle consisting of an electron dressed with a boson cloud. The quasiparticle may be heavier than a bare electron, and have unusual properties as a function of momentum. Two quasiparticles may have an attractive interaction, because their energy can be lowered by sharing the same boson cloud. This attractive interaction causes Cooper pairing and superconductivity.

We have developed a new, far more accurate and efficient method to calculate the properties of the polaron quasiparticle that is formed when the electron interacts with dynamical quantum phonons (in the Holstein model). The ground state energy we calculated is 10,000,000 times more accurate than any other published result, and was obtained using about a thousand times less computer resources. We have calculated the effective mass, properties at large momentum, correlation functions, excited state properties, and transport properties far from equilibrium. We have also obtained preliminary results for the binding energy of the bipolaron, which is the same as that of a Cooper pair in the low density limit (Pub. 55, 57, 58).

In a separate project, together with experimentalists in MST-10, we have investigated the dynamics of the destruction and reformation of the superconducting condensate in the YBCO high temperature superconductor when the sample is pumped by a short (femtosecond) optical pulse followed by either another optical pulse or a lower frequency (terahertz) probe. The pump breaks apart Cooper pairs and forms normal fluid quasiparticles. Several unexpected results have been obtained, including the fact that as the systems recovers, the extra normal fluid disappears before the superfluid reappears (Pub. 53, 56).

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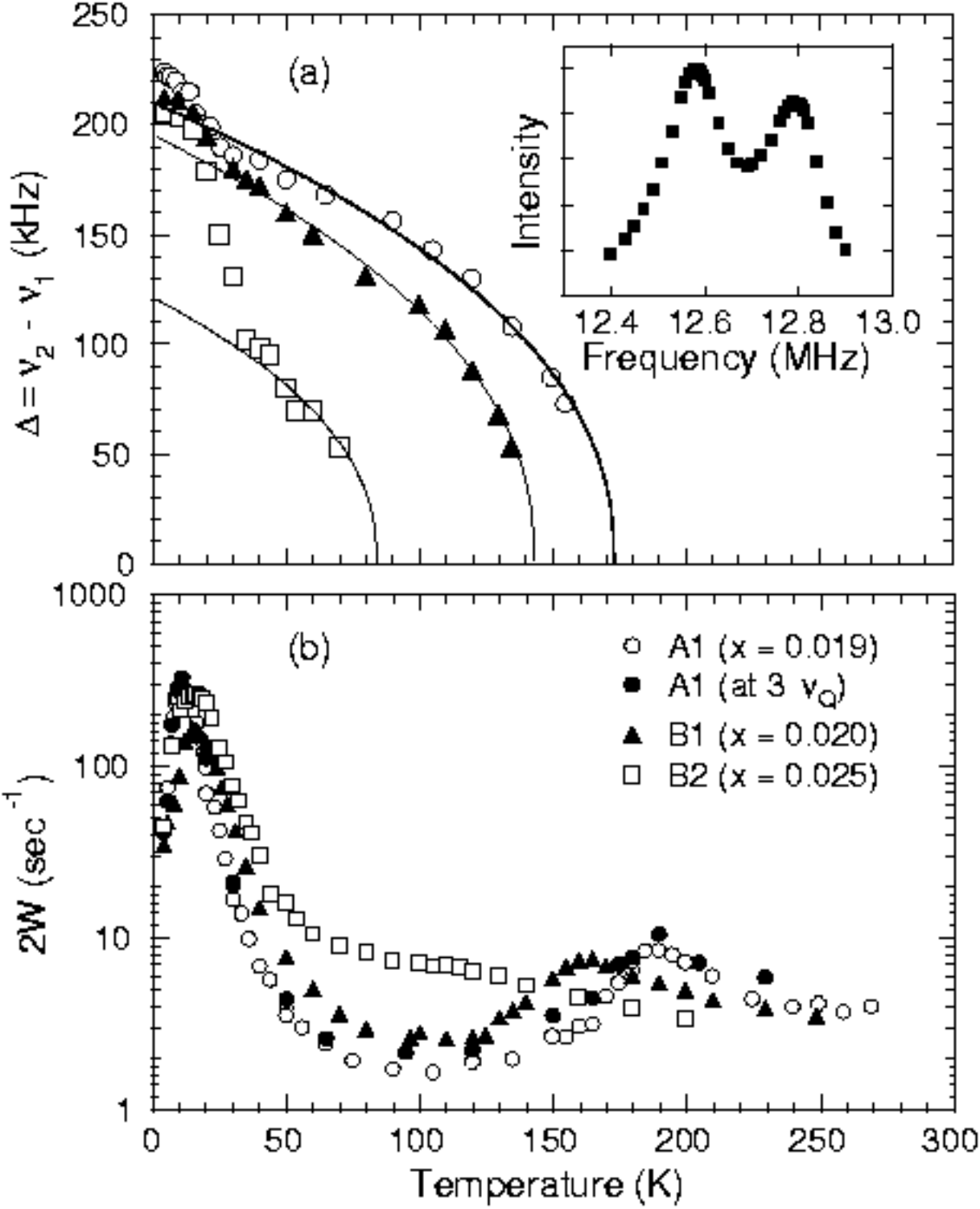
Figure 1: The magnetic properties of lightly Li-doped lanthanum cuprate, $La_2Cu_{1-x}Li_xO_4$, as a function of temperature is shown. The top panel shows the behavior of the static sublattice magnetization in the antiferromagnetically ordered state. The ordering temperature T_N is evident as the point where this magnetization vanishes with increasing temperature; this is suppressed by increasing doping (indicated by the values of x in the lower panel). Surprisingly, the magnitude of the magnetization is also suppressed, but recovers below ~30 K. The lower panel shows the ^{139}La spin lattice relaxation rate on a logarithmic scale. The very strong low temperature peak occurs at the temperature where the characteristic electron spin fluctuation frequency has slowed to the point where it matches the NMR frequency (~12 MHz). Essentially identical behavior is universally observed in antiferromagnetically ordered lanthanum cuprate, regardless of the nature or location of the dopant and for a wide range of doping concentration.

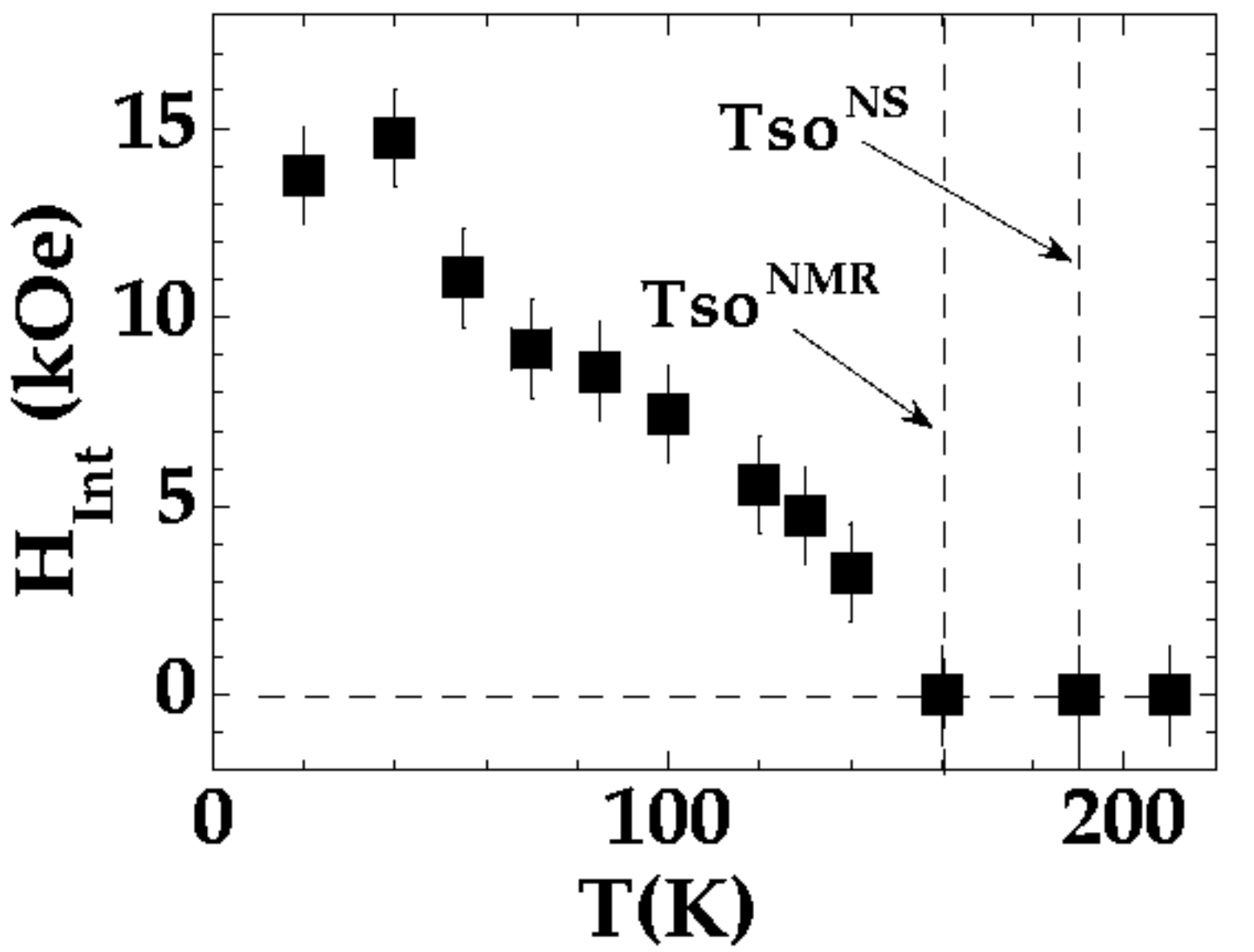
Figure 2: The temperature dependence of the static internal field due to antiferromagnetic order in charge-stripe ordered $La_{5/3}Sr_{1/3}NiO_4$ is shown. The antiferromagnetic ordering

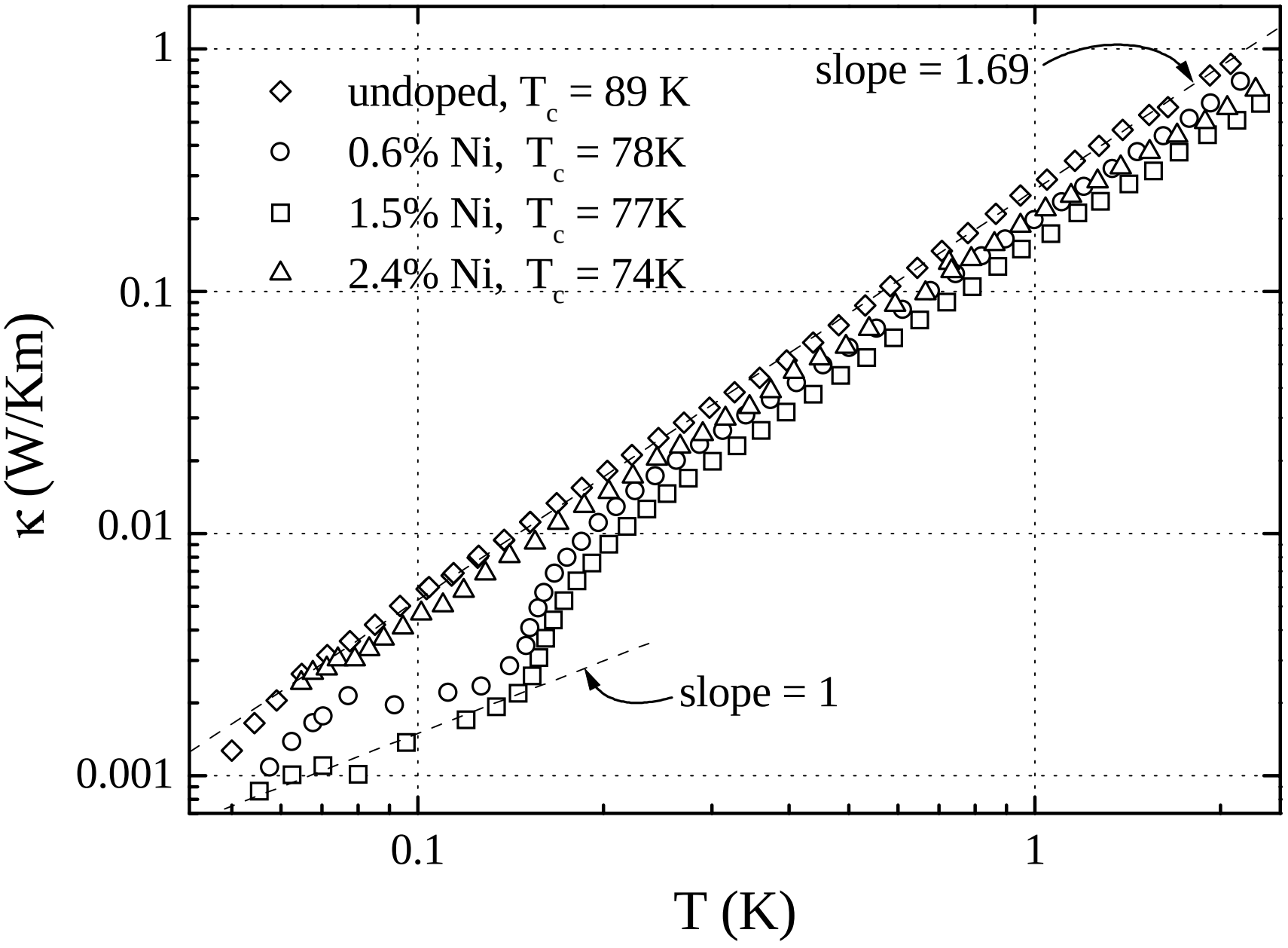
temperature TN = 160 K is evident as the temperature where the internal field vanishes with increasing temperature. An ordering temperature of 190 K was determined from neutron scattering measurements. The NMR measurements have a very slow characteristic measurement time (less than a microsecond), while the neutron scattering measurement corresponds to approximately 1-10 picoseconds. This sensitivity of ordering temperature to measurement time scale is a hallmark of glassy behavior, indicating the stripe solid is glass-like.

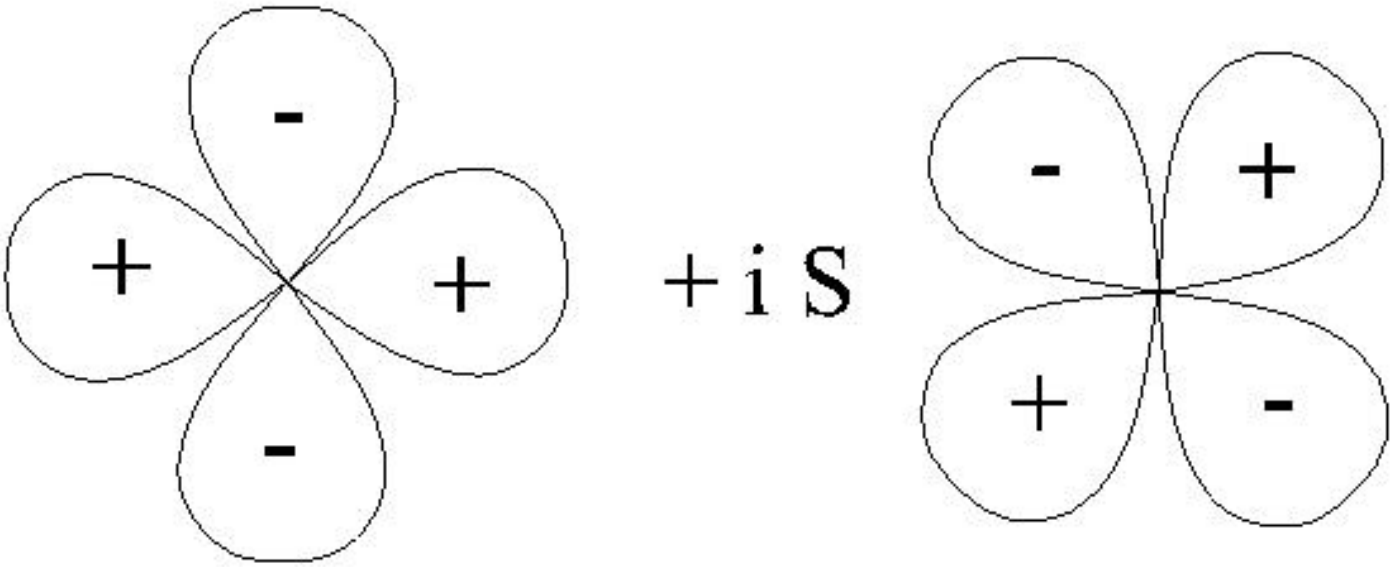
Figure 3: The low temperature thermal conductivity of several Bi2212 samples is shown. The thermal conductivity of two Ni-doped samples with T_c = 77-78 K show sharp drops at T^* = 200 mK. The sample is deep in the superconducting state, so this drop indicates a transition in superconducting order parameter. We propose the observed transition is from a d_{x2-y2} to a $d_{x2-y2} + id_{xy}$ state.

Figure 4: The figure illustrates a HTC (cuprate) superconductor that has a d_{x2-y2} order parameter in the bulk. The quasiparticle excitations are gapless in the (1,1) and symmetry related directions. A magnetic impurity has spin-orbit coupling to the electrons, and induces a d_{xy} superconducting order parameter component in its vicinity. The d_{xy} component has an imaginary coefficient, and the resultant total order parameter $|\Delta|$ is then gapped everywhere. This strongly reduces the population of thermally excited quasiparticles and hence the thermal conductivity, which is observed experimentally.


(a)
Intensity
12.4
12.6
12.8
13.0
Frequency (MHz)
$\Delta = \nu_2 - \nu_1$ (kHz)
250
200
150
100
50
0
(b)
A1 (x = 0.019)
A1 (at 3 ν_Q)
B1 (x = 0.020)
B2 (x = 0.025)
$2W$ (sec^{-1})
1000
100
10
1
0
50
100
150
200
250
300
Temperature (K)


15
10
5
0
H_{Int} (kOe)
0
100
200
T(K)
Tso^{NS}
Tso^{NMR}


1
0.1
0.01
0.001
κ (W/Km)
0.1
1
T (K)
undoped, T_c = 89 K
0.6% Ni, T_c = 78K
1.5% Ni, T_c = 77K
2.4% Ni, T_c = 74K
slope = 1.69
slope = 1



$d_{x^2-y^2}$ d_{xy}